

WRITTEN REPORT

The report by Crick et al. presents new information, in the form of sulfur isotope composition (^{33}S and ^{34}S) of volcanic eruption-sourced sulfate, on a number of volcanic events dated at about 74,000 years BP in two Antarctica ice cores. The sulfur isotope data show unambiguously that most of these events are explosive eruptions in the low latitudes that injected substantial amounts of sulfurous gases into the stratosphere (above the ozone layer). This is based on the findings in the early 2000s (e.g., Savarino et al. 2003; Baroni et al. 2007) that sulfate formed in the stratosphere from oxidation of certain sulfur species (mainly SO_2) possesses nonzero sulfur mass-independent fractionation (S-MIF) signatures. The experimental procedures used in this study, including ice core sampling (multiple samples in an event), sulfur isotope ratio measurement, and correction of isotope contribution from non-volcanic sulfate background in the calculation of S-MIF in ice core samples containing both background and volcanic sulfate, follow previously tested and verified methodology and, therefore, the data appear to be robust and of high quality.

From the point of view of identifying large, low latitude stratospheric eruptions in this time period, this study provides additional evidence, but reaches nearly the same conclusion as that by Svenssen et al. (2013), who examined the same group of volcanic events in Antarctica and Greenland cores. Svenssen et al. concluded, based on the simultaneous appearance of large sulfate signals in bipolar ice core records, that nine events (T1-T9) are potential candidates for the famed Toba eruption, dated by Ar isotope geochronometer to be around 73 ka. In this study, the S-MIF data verify that all, maybe with the exception of one, of these events are stratospheric. In addition, a careful examination of the isotope data indicates that locations of events T5-T9 may be extratropical, rather than low latitude.

Identification of the Toba eruption in ice cores

The above summary (by me) indicates that this study confirms results from previous work and adds significant new information. However, the main objective of this study (Lines 111-113) appears to be to identify, or to narrow the range of, the signal of the Toba eruption in ice cores. This intent is also suggested by the title of the paper (“the 74 ka Toba eruption”). Unfortunately, sulfur isotope signatures, similar to contemporaneous bipolar sulfate events, do not provide undisputable evidence of a specific eruption, even for Toba. Unlike tephra matching, unambiguous S-MIF data are not a “smoking gun”.

In this study, the identification of events T1, T2 and/or T3 as resulting from the Toba eruption is based on two pieces of evidence: the precise timing and the stratospheric nature of the events. Would there be other eruptions that meet these criteria? Or, in other words, can we eliminate the possibility that other eruptions left the volcanic sulfate of these events? The fact that at least three events (T1, T2 and T3) meet these criteria, with the small possibility that they were all left by the same Toba eruption, suggests we cannot be highly confident that the answer is yes. In fact, there are reasons to suspect that none of the candidate events is Toba.

The Toba eruption ejected a huge amount of materials – $3,800 \text{ km}^3$ DRE (Costa et al., 2014). This is more than three orders of magnitude that of the 1815 CE Tambora eruption ($\sim 1.2 \text{ km}^3$ DRE, Self et al., 2004). Estimates of the sulfur (aerosol) output of Toba are also several orders of magnitude larger than that of Tambora. (However, I would discount the aerosol

estimates from petrological/volcanological data or ice core data, as these rely on scaling factors (multipliers) that are poorly constrained.) One would expect that the Toba sulfate signal would be exponentially larger than that of Tambora in the same ice core. The volcanic sulfate flux/deposition of all of the three potential Toba events (Figure 2 and Table S1), except for T2 in EDML, is not overwhelmingly large: they are approximately 1-to-2 times that of Tambora. For T2, the much smaller flux (46.2 mg per square m) for EDC suggests that the flux (424) in EDML (about 9 times that of Tambora) may be an outlier. The sulfate flux data of these events in Greenland cores (Svensenn et al., 2013) are also approximately 1-to-2 times that of Tambora. If the Toba eruption resulted in one of the three events, why is its sulfate flux so much smaller than what would be expected? Toba would have to be an exceptionally sulfur-poor eruption to leave one of the three volcanic sulfate signals in ice cores.

In a recently published Holocene volcanic record (Cole-Dai et al., 2021) from the West Antarctica WDC core, ten eruptions in the Holocene are found in the range of 1-2 times of Tambora flux. This suggests that events similar to T1, T2 and T3 in magnitude are not uncommon – they occur relatively frequently, at about one per millennium. The magnitude of ice core signals of T1, T2 and T3 suggests that the eruptions they represent are not at the Toba scale.

The much-smaller-than-expected sulfate signal could be the enigma for identifying Toba in ice cores in ice cores.

Estimating eruption plume altitude

The authors of this discussion paper use the extreme cap-delta-33-S values of T1 and T2 to infer that the plume altitude of the eruption clouds must be exceptional high. In fact, they estimate the plume altitude to be at least 45 km for T1 and T2 (Lines 329-330). The estimate is derived or extrapolated from an empirical quantitative relationship between cap-delta-33-S and plume altitude (Figure 6). I question the validity of the extrapolation for two reasons. First, the quantitative relationship is based on four eruptions (Agung, Pinatubo, Samalas and Tambora) or data points with very large uncertainties. The maximum magnitude of cap-delta-33-S for a volcanic event depends strongly on the sampling resolution during the event, as the value of cap-delta-33-S evolves from positive to negative. This is analogous to peak height measurement dependent on sampling resolution during the peak. As a result, I suspect that the uncertainties for maximum cap-delta-33-S values are larger than seen in Figure 6.

Second, the authors cite the study of Lin et al. (2018) to support their proposal that the magnitude of cap-delta-33-S in volcanic sulfate is dependent on the altitude in the stratosphere where the sulfate is formed. I read the Lin et al. paper and have a different understanding of the conclusion regarding that relationship between cap-delta-33-S and altitude. First, Lin et al. measure S-MIF in tropospheric sulfate; therefore, the relationship they describe is for tropospheric altitudes. I think it is quite a stretch to argue that such a relationship could be extrapolated into the stratosphere. Second, Lin et al. explained that the relationship is the result of downward transport of stratospheric sulfate with non-zero cap-delta-33-S; this transport from the stratosphere is supported by an altitude-dependent trend of 35S which is only produced in the stratosphere or above. I think it is on a very shaky ground to use the altitude-S-MIF relationship found by Lin et al. to justify a similar relationship for volcanic sulfate in the stratosphere and to estimate the plume altitude of the volcanic eruption. In my view, interpretation of the volcanic S-MIF magnitude is premature; much more research is required to understand the significance of the volcanic S-MIF magnitude.

Recommendation

I would recommend that the paper be revised to (1) de-escalate the certainty that the fallout of the Toba eruption is among the volcanic signals examined, in a fashion similar to what Svenssen et al. did in reaching conclusions regarding Toba identification, and (2) reconsider including estimating eruption plume altitude from the S-MIF data.